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(54) **POLYESTER COMPOSITION AND MOLDINGS THEREOF.**

(57) A polyester composition comprising (a) an aromatic polyester having a relative viscosity of 1.2 to 20, (b) an aromatic polycarbonate having a number-average molecular weight of 10,000 to 80,000, and (c) a glycidyl group-containing copolymer of 0.1 to 100 in melt index containing as major components an α-olefin and a glycidyl ester of an α, β-unsaturated acid. This composition possesses excellent fluidity and residence stability upon molding, and excellent mechanical properties, particularly impact resistance, thus being useful as electric and electronic device parts and automobile parts. This composition can be molded into various moldings in a conventional manner.

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SPECIFICATION

POLYESTER COMPOSITIONS AND MOLDED ARTICLES THEREFROM

Field of Art

The subject invention relates to the polyester compositions being superior in flow property and melt stability on molding, as well as in mechanical properties particularly in impact resistance and hot-air aging deterioration resistance, and to the molded articles therefrom.

Background Art

Having superior characteristics, aromatic polyesters that are represented by polyethyelene terephthalate and polybutylene terephthalates are extensively used for manufacturing electric, electronic and automotive parts for example. However, their uses do not increase because of their low impact resistances. Therefore, it has hitherto been proposed to blend therein various kinds of polymers such as butadiene rubbers and acrylic elastomers. Among these blending methods, those with the copolymers being composed of such monomers as α-olefin and glycidyl ester of α,β-ethylenically unsaturated acid that are mentioned in

Japanese Laid-Open Patent Publication (Kokai) No. 32045/1977 and the U.S. Patent No. 4461871 are comparatively superior to the others in improving effect of impact resistance and melt stability on molding. Nevertheless, in these methods, there is a problem that molded articles deteriorate in the impact resistance by hot-air aging in an oven. Further, the blendings of aromatic polycarbonates and acrylic elastomers to polybutylene terephthalates that are proposed in Japanese Laid-Open Patent Publication (Kokai) 500870/1980 are not satisfiable in impact resistance despite small increase and inferior in melt stability. The method soluing the above-mentioned problems has not been found.

Disclosure of the Invention

The object of the subject invention is to provide the aromatic polyester compositions being superior in flow property and melt stability on molding, as well as in mechanical properties particularly in impact resistance and hot-air aging resistance, and being useful for manufacturing lectric, electronic and automotive parts, and their molded articles.

The object is achieved by a polyester composition being composed of:

(a) an aromatic polyester having the relative viscosity of 1.2 to 2.0,

(b) an aromatic polycarbonate having the number average molecular weight of 10,000 to 80,000, and

(c) a glycidylgroup containing copolymer consisting essentially of α-olefin and glycidyl ester of α,β-ethylenically unsaturated carboxylic acid and having the melt index of 0.1 to 100,

wherein components (a) and (b) are present in a weight ratio in the range of between 99/1 and 1/99 respectively and the component (c) is present in an amount of from 1 to 80 parts by weight per the total 100 parts by weight of components (a) and (b).

The object is preferably attained by the above compositions containing the ethylene based copolymer being composed of ethylene and α-olefin having 3 to 10 carbon atoms also.

The Best Forms to Practice the Invention

The subject invention will be described in further details hereinafter.

The resins being used according to the subject invention contain aromatic polyesters and aromatic polycarbonates.

The former are the polymers or copolymers having

aromatic rings in chains and prepared by condensing mainly aromatic dicarboxylic acid (or their ester forming derivative) and diol (or their ester forming derivative).

The above-mentioned aromatic dicarboxylic acids include terephthalic acid, isophthalic acid, ortho phthalic acid, 2,6-naphthalenedicarboxylic acid, 1,5-naphthalenedicarboxylic acids, bis(p-carboxyphenyl)methane, anthracenedicarboxylic acid, 4,4'-diphenyldicarboxylic acid, diphenyletherdicarboxylic acid, 1,2-bis(4-carboxyphenoxy)-ethane and so forth and ester forming derivatives thereof.

The above-mentioned aromatic dicarboxylic acid may be replaced with aliphatic dicarboxylic acids such as adipic acid, sebacic acid, azelaic acid, dodecanedionic acid and so forth. Alicyclic dicarboxylic acids such as 1,3-cyclohexanedicarboxylic acid, 1,4-cyclohexanedicarboxylic and so forth and ester forming derivatives thereof, provided they are present in an amount of less than 40 mol percents based on the total acid component.

The diols include the aliphatic diols having 2 to 10 carbon atoms such as ethylene glycol, propylene glycol, 1,4-butane diol, neopentyl glycol, 1,5-pentane diol, 1,6-hexane diol, decamethylene glycol, cyclohexanedimethanol

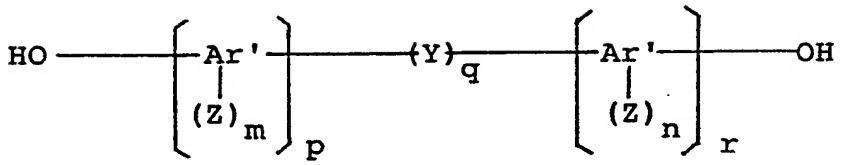
and their mixtures. Further the small amount of long-chain glycols having molecular weights in the range of 400 to 6,000 such as polyethylene glycol, poly-1,3-propylene glycol, polytetramethylene glycol and so forth as well as their mixtures can be copolymerized.

Preferable aromatic polyesters according to the subject invention are polyethylene terephthalate, polypropylene terephthalate, polybutylene terephthalate, polyhexamethylene terephthalate, polycyclohexylenedimethylene terephthalate, polyethylene-2,6-nathalate and so forth. Most preferable thereamong are polybutylene terephthalate that has exellent mechanical strength.

The aromatic polyesters should preferably have a relative viscosity of 1.2 to 2.0, more preferably of 1.4 to 1.8 as measured by a 0.5 percent orthochlorophenol solutions at 25°C. Insufficient mechanical strength is developed or no good luster surface moldings are obtained when they are less than 1.2 or more than 2.0 respectively.

The aromatic polycarbonates according to to the subject invention are preperable by ester exchange or phosgene methods using dihydric phenol or its derivative.

The dihydric phenols are represented by the following formula:



wherein Ar' denotes an aromatic group such as phenylene, biphenylene and naphthylene; Z denotes an alkyl group such as methyl and ethyl, a halogenated alkyl group, an aryl group such as phenyl and naphthyl, a halogenated aryl group, aralkyl group such as benzyl and phenylethyl, a halogenated aralkyl group, or an alicyclic group; Y denotes an alkylene group such as methylene and ethylene, an alkylidene group such as ethylidene and isopropylidene, a tertiary amino group, O, S, SO, SO_2, CO or an amide group; m and n are integers from 0 to 4; p is integer at least 1; q is 0 or 1; or r is 0 or positive integer. When q is o, r is o.

Illustrative of the dihydric phenols are;

bis(4-hydroxyphenyl)-methane;

1,1-bis(4-hydroxyphenyl)-ethane;

1,2-bis(4-hydroxyphenyl)-ethane;

2,2-bis(4-hydroxyphenyl)-propane;

1,1-bis(4-hydroxyphenyl)-propane;

2,2-bis(4-hydroxy-3-chlorophenyl)-propane;

2,2-bis(4-hydroxy-3,5-dichlorophenyl)-propane;

2,2-bis(4-hydroxy-3-bromophenyl)-propane;

2,2-bis(4-hydroxy-3,5-dibromophenyl)-propane;

2,2-bis(4-hydroxy-3-methylphenyl)-propane;

2,2-bis(4-hydroxy-3-methoxyphenyl)-propane;

1,4-bis(4-hydroxypyenyl)-cyclohexane;

1,1-bis(4-hyroxyphenyl)-cyclohexane;

1,2-bis(4-hydroxyphenyl)-ethylene;

1,4-bis(4-hydroxyphenyl)-benzene;

bis(4-hydroxyphenyl)-phenylmethane; bis(4-hydroxyphenyl)-diphenylmethane;

1,1-bis(4-hydroxyphenyl)-2,2,2-trichloroethane;

bis(4-hydroxyphenyl)-ketone; bis(4-hydroxyphenyl)-sulfide;

bis(4-hydroxyphenyl)-sulfone; 4,4'-dihydroxydiphenyl ether;

4,4'-dihydroxybiphenyl; 3,3'-dihydroxybiphenyl;

hydroquinone; resorcinol; 2,6-dihydroxynaphthalene;

2,7-dihydroxynathalenes; phenophthalein; and so forth.

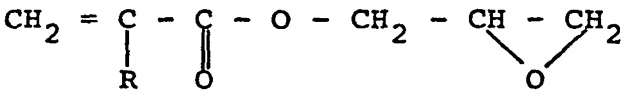
Thereamong bis(4-hydroxyphenyl)alkane is preferable and 2,2-bis(4-hydroxyphenyl)-propane is especially preferable. More than two dihydric phenols are usable in combination. They may be used together with the small quantities of alicyclic diols such as 1,4-cyclohexane diol, aliphatic diols such as 1,6-hexane diol, aromatic group containing aliphathic diols such as p-xylene glycol and so forth or can be end-capped by monohydric phenols such as phenol and p-tert-butylphenol..

The aromatic polycarbonate has the number-average

molecular weight of 10,000 to 80,000, preferably of 15,000 to 40,000. The compositions can neither obtain enough mechanical properties nor hot-air aging resistance if it is less than 10,000 or inferior in moldability and mechanical properties if they are more than 80,000.

The aromatic polyester and aromatic polycarbonate according to the subject invention are present in a weight ratio in the range between 99/1 and 1/99, preferably between 80/20 and 20/80 respectively. On the contrary the synergitic effect for the increase in impact resistance of polyester compositions and the resulting increase in hot-air aging resistance are small except the above-mentioned ranges.

Of the glycidyl-group containing copolymers consisting essentially of α-olefin and glycidyl ester of α,β-ethylenically unsaturated carboxylic acid, the former include ethylene, propylene, butene-1 and so forth, of which ethylene is preferable. The latter compound represented by the following general formula:



wherein R denotes a hydrogen atom, a lower alkyl group or a

glycidyl-group substituted lower alkyl group. Glycidyl acrylate, glycidyl methacrylate, glycidyl ethacrylate and glycidyl itaconate are the examples. Thereof, glycidyl m thacrylate is preferable. The copolymers advantageously have a glycidyl unit of α,ß-ethylenically unsaturated carboxylic acid content in the range from 0.5 to 40 percent, preferably from 1 to 30 percent, more preferably from 2 to 20 percent by weight. If the contents are less than 0.2 percent or more than 40 percent, the copolymers do not satisfactorily increase in impact resitance or decrease in molding flowability respectively. Glycidyl esters of α,ß-ethylenically unsturated carboxylic acid can be copolymerized by standard copolymerization or graft reaction. Further, less than 40 percents by weight of at least one unsaturated monomers such as: vinyl ethers; vinyl acetate, propione and other vinyl esters; methyl, ethyl, propyl and butyl and other esters of acrylic or methacrylic acid; acrylonitrile; styrene; and carbon monoxide may be copolymerized with the above copolymers.

The MI (Melt Index) of glycidyl-group containing copolymer is in the range of between 0.1 and 100, preferably between 0.5 and 30, wherein the value of MI is measured at 190°C according to ASTM D-1238 and the unit is gr./10 min. If the MI is less than 0.1 gr./min. or more than 100

gr/10min. increase is small in impact resistance.

Preferable among glycidyl-group containing copolymers are ethylene / glycidyl methacrylate, ethylene / vinyl acetate / glycidyl methacrylate, ethylene / carbon monoxide / glycidyl methacrylate, ethylene / glycidyl acrylate, and ethylene / glycidyl acrylate / vinyl acetate copolymers. Among them ethylene / glycidyl methacrylate copolymer is more preferable.

The glycidyl-group containing copolymers according to the subject invention are present in an amount of from 1 to 80 parts by weight, preferably of 5 to 50 parts by weight, per the total 100 parts by weight of aromatic polyester and aromatic polycarbonate. If the amount is less than 1 part or more than 80 parts, polyester compositions do not satisfactorily increase in impact resitance or aromatic polyesters deteriorate in mechanical properties.

The compositions according to the subject invention further increase in impact resistance when containing the ethylene based copolymer being composed of ethylene and α-olefin having 3 to 10 carbon atoms and/or the diene copolymer being composed of ethylene, α-olefin having 3 to 10 carbon atoms and unconjugated diene.

The above-mentioned α-olefins include propylene, butene-1, pentene-1, 3-methylpentene-1, octacene-1, decene-1

and so forth. Thereof, propylene and butene-1 are preferable and more than two usable in combination.

The unconjugated dienes include various kinds of norbonene compounds, dicyclopentadiene compounds, tetrahydroindene compounds, 1,4-hexadiene and so forth. Preferable thereamong are 5-ethylidene-2-norbonene, dicyclopentadiene and 1,4-hexadiene.

The molar ratios of ethylene to α-olefin in the ethylene based copolymers are in the range between 40/60 and 99/1, preferably between 70/30 and 95/5, and in the diene copolymers the copolymerized ratios of α-olefin and unconjugated diene are in an amount of from 5 to 80 mol percent, preferably from 10 to 60 mol percent and from 0.1 to 20 mol percent, preferably from 0.5 to 10 mol percent, respectively.

The ethylene based copolymer and / or the diene copolymers are present in an amount of from 1 to 50 parts by weight, preferably from 5 to 40 parts by weight, per the total 100 parts by weight of aromatic polyesters and aromatic polycarbonates.

The compositions according to the subject invention can be increased in stiffness by further adding inorganic fillers. This addition generally causes the decrease in impact resistance. It is however small in the case of the

compositions according to the subject invention.

Among the inorganic fillers according to the subject invention, fibrous and granular ones as well as their mixtures can be mentioned. The fibrous ones include glass, silas glass, almina, silicon carbide, ceramic, asbestos, gypsum, metal (e.g. stainless steel) and other inorganic and carbon fibers. The granular ones, on the other hand, include wollastonite, sericite, kaolin, mica, clay, bentonite, asbestos, talc, alumina silicate and other silicates; metal oxides such as alumina as well as silica, magnesium oxide, zirconium oxide and titanium oxide; carbonates such as calcium carbonate and magnesium carbonate as well as dolomite; sulfates such as calcium sulfate and barium sulfate; glass beads; boron nitride; silicone carbide; sialon. They are permitted to be hollow (e.g. hollow glass fiber, glass microballoon, silas balloon, carbon balloon, etc.). Preferable thereamong are glass fibers, carbon fibers, metal fibers, potassium titanate whisker, glass flakes, glass beads, wollastonite, mica, talc, clay, titanium oxide, aluminum oxide, calcium carbonate and barium sulfate. Particularily thereamong glass fiber is more preferable. The inorganic fillers should preferably be treated with silane, titanate or another conventional coupling agent, and glass fibers with

an conventional converging agent such as epoxy resin and vinyl acetate resin.

The inorganic fillers are be added at the ratios by weight of 3 to 100 parts, preferably of 5 to 80 parts, per the total 100 parts by weight of aromatic polyesters and aromatic polycarbonates.

The compositions according to the subject invention can be increased in impact resistance by adding the compounds for promoting the reaction between epoxy compounds and carboxylic acids. They include triphenyl amine, 2,4,6-tris(dimethylaminomethyl)phenol and other tertiary amines; triphenyl and trisodecyl phosphites and other phosphite esters; triphenylallylphosphonium bromide and other phosphonium compounds; triphenylphosphine and other tertiary phosphines; lithium, calcium and other metal stearates; sodium dodecylbenzenesulfonate and sodium 3,5-dicarbomethoxybenzenesulfonate and other metal sulfonates; sodium lauryl sulfate and other organic sulfate salts, and so forth. Their additon should be made at the ratios of 0.001 to 5 parts by weight to 100 parts by weight of aromatic polyesters.

The compositions according to the subject invention permit the addition of such quantities as not obstructing its object of more than one being selected from fibrous and

granular fillers and reinforcements, antioxidants, heat stabilizers, ultraviolet ray-absorbents, lubricants, mold releasing agents, colorants including dyes and pigments, flame retardants and flame redarding assistants, antistatic agents, crystallization promotors, and other additives as well as of the small quantities of one or more than two being selected out of thermoplastic resins, thermosetting resins and thermoplastic elastomers.

The processes for producing the compositions according to the subject invention are not limited. However, preferable thereamong are to melt-extrud, by using an extruder, the dry-blendings of aromatic polyesters, aromatic polycarbonates, glycidyl group containing copolymers and, if necessary, other additives.

The resin compositions of the subject invention can be molded according to conventional methods such as injection molding, extrusion molding, and molded articles therefrom show excellent properties.

The effects of the subject invention is illustrated below in detail with reference to Examples. The Examples are by way of illustration and not by way of limitation.

Examples 1 - 8:

The dry-blendings of the polybutylene terephthalate (PBT) having a relative viscosity of 1.56 and the amounts

shown in Table 1 of the ones selected as shown therein of aromatic polycarbonates derived from 2,2'-bis(4-hydroxyphenyl)propane and glycidyl-group containing copolymers were melt-extruded and pelletized by using a screw extruder set at 250°C. The melting viscosity of these pellets was measured at 250°C by using a koka-type flow tester. Subsequently they were molded into ASTM No. 1 dumbbells and 1/4-in. wide Izod impact testpieces using a 5-oz. screw in-line type injection molding machine. While injection molding, the minimum injiction (lower-limit molding) pressure required for mold charge was measured. The testpieces underwent tensile and notched Izod impact tests. Further they were kept heat-treated in an oven at 150°C for 500 hours and subjected to tensile and notched Izod impact tests. Their results are shown in the Table 1.

Table 1

		PBT (Part by weight)	Aromatic polycarbonates		Glycidyl-group containing copolymers		Mechanical properties						Melting viscosity (poise)	Lower limit Pressure (kg/cm^2)
							Not treated			Heat-treated				
			Kinds	Amounts (parts by weight)	Kinds	Amounts (parts by weight)	Tensile strength (kg/cm^2)	Elongation at break (%)	Izod impact strength (kg.cm/cm-notch)	Tensile strength (kg/cm^2)	Elongation at break (%)	Izod impact strength (kg.cm/cm-notch)		
Examples	1	90	A	10	E	25	380	195	46	386	96	40	5300	43
	2	70	A	30	E	25	383	200 <	59	391	151	53	5500	44
	3	50	A	50	E	25	398	200 <	70	409	159	61	7100	50
	4	30	A	70	E	25	404	200 <	65	413	150	55	8000	53
	5	10	A	90	E	25	410	181	47	411	93	39	7800	52
	6	70	B	30	E	25	380	190	41	389	112	33	6200	47
	7	70	A	30	E	5	435	200 <	17	441	140	15	4900	39
	8	70	A	30	E	40	339	200 <	62	347	132	50	8500	54
Comparative Examples	1	100	-	-	-	-	569	120	3.2	572	15	1.8	3100	34
	2	100	-	-	E	25	388	184	35	394	25	5.5	5200	43
	3	70	A	30	-	-	577	115	4.3	586	32	3.1	4200	37
	4	50	A	50	-	-	592	73	10	601	27	5.3	6500	48
	5	70	A	30	E	90	329	132	45	332	59	27	14300	73
	6	98	A	2	E	25	375	195	37	391	51	8.1	5300	43
	7	70	C	30	E	25	371	57	21	382	20	12	11800	63
	8	70	D	30	E	25	362	63	25	368	27	11	4500	38
	9	70	A	30	F	25	364	105	22	369	39	8.5	4300	37
	10	70	A	30	G	25	365	51	16	357	25	7.2	7500	51
	11	70	A	30	H	25	357	44	13	354	11	6.3	13200	69
	12	70	A	30	I	25	370	56	10	365	20	4.5	4200	37

Note*: aromatic polycarbonates

A: number average molecular weight = 25,000

B: number average molecular weight = 62,000

C: number average molecular weight = 95,000

D: number average molecular weight = 9,000

Note**: specifications of glycidyl-group containing copolymers

Codes	Ethylene/glycidyl methacrylate ratio by weight	M I gr./10 min.
E	90/10	3.0
F	90/10	120
G	90/10	0.05
H	55/45	3.8
I	99.8/0.2	2.5

The results in the Table 1indicates that resin compositions increase in impact resistance and their molded articles are superior in hot-air aging resistance if they contain both polycarbonates and glycidyl-group containing copolymers according to the subject invention.

Example 9 - 12:

Evaluation was carried out in the same manner as

Example 3 except that 15 of the 25 parts by weight of the glycidyl-group containing copolymer E was replaced with the ethylene based copolymer or diene copolymers shown in Table 2. Their results are shown in Table 2.

Table 2

		Kinds of ethylene based co-polymer or diene copolymer *	Mechanical properties						Melting viscosity (poise)	Lower limit pressure (kg/cm^2)
			Not treated			Heat-treated				
			Tensile strength (kg/cm^2)	Elongation at break (%)	Izod impact strength (kg.cm/cm notch	Tensile strength (kg/cm^2)	Elongation at break (%)	Izod Impact strength (kg.cm/cm notch		
Examples	9	J	393	200<	83	398	175	76	6500	48
	10	K	395	200<	87	403	183	80	6300	47
	11	L	389	200<	82	394	166	73	6600	48
	12	M	390	200<	88	400	171	76	6600	48

Note*: ethylene based copolymer or diene copolymer

J: ethylene/propylene (80/20 molar ratio) copolymer MI = 1.5

K: ethylene/butene-1 (90/10 molar ratio) copolymer MI = 3

L: ethylene/propylene/dicyclopentadiene (70/28/2 molar ratio) copolymer MI = 0.8

M: ethylene/butene-1/5-ethylidene-2-norbonene (88/10/2 molar ratio) copolymer MI = 1

The results indicate that the resin compositions according to the subject invention further increase in impact strength and molding flowability when containing ethylene based copolymer or diene copolymer.

Examples 13 - 14:

The dry-blendings obtained by adding such amounts as shown in Table 3 of glass fibers (3mm-long chopped strands) to the polybutylene terephthalate (PBT) having a relative viscosity of 1.45, the aromatic polycarbonate A and glycidyl-group containing copolymer E used in Examples 1 were evaluated in the same manner as it except the measuring melting viscosity and lower limit molding pressure. The results of these evaluation are also shown in the Table 3.

Table 3

		PBT (parts by weight)	Aromatic poly-carbonate A (parts by weight)	Glycidyl-group containing copolymer E (parts by weight)	Inorganic filler (parts by weight)	Mechanical properties					
						Not treated			Heat-treated		
						Tensile strength (kg/cm^2)	Elongation at break (%)	Izod impact strength (kg.cm/cm-notch)	Tensile strength (kg/cm^2)	Elongation at break (%)	Izod impact strength (kg.cm/cm-notch)
Examples	13	50	50	20	10	740	8.3	27	735	7.9	25
	14	50	50	20	20	870	6.6	18	860	6.1	16
Comparative Examples	13	60	60	-	10	735	5.2	3.4	705	3.8	3.2
	14	60	60	-	20	860	4.1	4.0	835	3.4	3.6
	15	120	-	20	20	860	5.7	9.5	840	4.2	5.8

The results indicate that the compositions according to the subject invention increase in impact resistance and hot-air aging resistance as well as in stiffness if inorganic fillers are added besides.

Industrial Applicability of the Invention

The polyester compositions according to the subject invention are useful for manufacturing electric, electronic and automative parts.

Claims

1. The polyester composition being composed of:

(a) an aromatic polyesters having the relative viscosities of 1.2 to 2.0,

(b) an aromatic polycarbonates having the number average molecular weights of 10,000 to 80,000, and

(c) a glycidyl-group containing copolymer consisting essentially of α-olefin and glycidyl ester of α,β-ethylenically unsaturated carboxylic acid and having the melt index of 0.1 to 100, wherein components (a) and (b) are present in a weight ratio in the range of between 99/1 and 1/99 respectively and the component (c) is present in an amount of from 1 to 80 parts by weight per the total 100 parts by weight of components (a) and (b).

2. The composition, as claimed in Claim 1, wherein said aromatic polyester and aromatic polycarbonate are present in a weight ratio in the range of between 80/20 and 20/80 respectively.

3. The composition, as claimed in Claim 1, wherein glycidyl-group containing copolymer is present in an amount of from 5 to 50 parts by weight per the total 100 parts by weight of said aromatic polyester and aromatic polycarbonate.

4. The composition, as claimed in Claim 1, wherein the aromatic polyester is polyalkylene terephthalate.

5. The composition, as claimed in Claim 4, wherein the polyalkylene terephthalate is polybutylene terephthalate.

6. The composition, as claimed in Claim 1, wherein the aromatic polycarbonate is bis(4-hydroxyphenyl)alkane polycarbonate.

7. The composition, as claimed in Claim 6, wherein bis(4-hydroxyphenyl)alkane is 2,2-bis(4-hydroxyphenyl)propane.

8. The composition, as claimed in Claim 1, wherein said aromatic polycarbonate has the number average molecular weight of 15,000 to 40,000.

9. The composition, as claimed in Claim 1, wherein the glycidyl ester of α,β-ethylenically unstruated carboxylic acid of said glycidyl-group containing copolyemer is selected from the group of glycidyl methacrylate and glycidyl acrylate.

10. The composition, as claimed in Claim 1, wherein α-olefinis is ethylene.

11. The composition, as claimed in Claim 9 or 10, wherein the glycidyl-group containing copolymer is selected from the group of ethylene / glycidyl methacrylate

copolymer and ethylene / glycidyl methacrylate / vinyl acetate copolymer.

12. The composition, as claimed in Claim 1, wherein the copolymerized amount of the glycidyl ester of α,β-ethylenically unsaturated carboxylic acid in the glycidyl-group containing copolymer ranges from 0.5 to 40 percents by weight.

13. The composition, as claimed in Claim 1, further comprising the ethylene based copolymer being composed of ethylene and α-olefins having 3 to 10 carbon atoms in an amount of from 1 to 50 parts by weight per the total 100 parts by weight of said aromatic polyester and aromatic polycarbonate.

14. The composition, as claimed in Claim 1, fruther comprising the diene copolymer being composed of ethylene, the α-olefin having 3 to 10 carbon atoms and unconjugated diene in an amount of from 1 to 50 parts by weight per the total 100 parts by weight of said aromatic polyester and aromatic polycarbonate.

15. The composition, as claimed in Claim 13, wherein said ethylene based copolymer is selected from the group of ethylene/propylene copolymer, ethylene/butene-1 copolymer and ethylene/propylene/butene-1 copolymer.

16. The composition, as claimed in Claim 14, wherein said

diene copolymer contains more than one being selected from propylene and butene-1 as α-olefin and more than one being selected from dicyclopentadiene, 5-ethylidene-2-norbonene and 1,4-hexadiene as unconjugated diene.

17. The composition, as claimed in Claim 1, further comprising up to 50 percent by weight of inorganic filler.

18. The composition, as claimed in Claim 17, wherein said inorganic filler is selected from the group of glass fibers, carbon fibers, metal fibers, potassium titanate whisker, glass flakes, glass beads, wollastonite, mica, talc, clay, titanium oxide, aluminium oxide, calcium carbonate and barium sulfate.

19. The injection molded articles of the polyester composition being composed of:

(a) an aromatic polyester having the relative viscosity of 1.2 to 2.0,

(b) an aromatic polycarbonate having the number average molecular weight of 10,000 to 80,000, and

(c) a glycidyl-group containing copolymer consisting essentially of α-olefin and glycidyl ester of α,ß-ethylenically unsaturated carboxylic acid and having the melt index of 0.1 to 100, components

(a) and (b) are present in a weight ratio in the range of between 99/1 and and 1/99 by weight respectively and the component (c) is present in an amount of from 1 to 80 parts by weight per the total 100 parts by weight of components (a) and (b).

20. The extrusion molded articles of the polyester composition being composed of:

(a) an aromatic polyester having the relative viscosity of 1.2 to 2.0,

(b) an aromatic polycarbonate having the number average molecular weight of 10,000 to 80,000, and

(c) a glycidyl-group containing copolymer consisting essentially of α-olefin and glycidyl ester of α,β-ethylenically unsaturated carboxylic acid and having the melt index of 0.1 to 100, wherein components (a) and (b) are present in a weight ratio in th range of between 99/1 and 1/99 and the component (c) is present in an amount of 1 to 80 parts by weight per the total 100 parts by weight of components (a) and (b).

INTERNATIONAL SEARCH REPORT

International Application No. PCT/JP85/00242

I. CLASSIFICATION OF SUBJECT MATTER (if several classification symbols apply, indicate all) [3]

According to International Patent Classification (IPC) or to both National Classification and IPC

Int. Cl[4] C08L67/02, C08L69/00, C08L63/00

II. FIELDS SEARCHED

Minimum Documentation Searched [4]

Classification System	Classification Symbols
IPC	C08L67/00-67/02, C08L69/00, C08L63/00

Documentation Searched other than Minimum Documentation to the Extent that such Documents are Included in the Fields Searched [5]

III. DOCUMENTS CONSIDERED TO BE RELEVANT [14]

Category*	Citation of Document, [16] with indication, where appropriate, of the relevant passages [17]	Relevant to Claim No. [18]
X	US, A, 4,172,859 (E. I. du Pont de Nemours and Company), 30 October 1979 (30. 10. 79) & DE, A, 2,622,876 & FR, B, 2,311,808 & GB, A, 1,552,637 & IT, A, 1,061,452 & NL, A, 7,605,494 & JP, A, 51-144452	1 - 12, 17 - 20
X	JP, A, 58-91759 (Toray Industries, Inc.), 31 May 1983 (31. 05. 83), Page 3, lower left column, lines 4 to 15 (Family : none)	1, 3, 4, 9 - 12 17 - 20
X	JP, A, 58-71941 (Toray Industries, Inc.), 28 April 1983 (28. 04. 83), Page 4, upper left column, lines 10 to 20 (Family : none)	17, 18
Y	US, A, 4,172,859 (E. I. du Pont de Nemours and Company), 30 October 1979 (30. 10. 79) & DE, A, 2,622,876 & FR, B, 2,311,808 & GB, A, 1,552,637 & IT, A, 1,061,452 & NL, A, 7,605,494 & JP, A, 51-144452	13 - 16
Y	JP, A, 58-91759 (Toray Industries, Inc.), 31 May 1983 (31. 05. 83) (Family : none)	2, 6-8, 13 - 16

* Special categories of cited documents: [15]

"A" document defining the general state of the art which is not considered to be of particular relevance

"E" earlier document but published on or after the international filing date

"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)

"O" document referring to an oral disclosure, use, exhibition or other means

"P" document published prior to the international filing date but later than the priority date claimed

"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step

"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art

"&" document member of the same patent family

IV. CERTIFICATION

Date of the Actual Completion of the International Search [3]	Date of Mailing of this International Search Report [3]
July 4, 1985 (04. 07. 85)	July 15, 1985 (15. 07. 85)
International Searching Authority [1]	**Signature of Authorized Officer [20]**
Japanese Patent Office	

FURTHER INFORMATION CONTINUED FROM THE SECOND SHEET

Y	JP, A, 58-71941 (Toray Industries, Inc.), 28 April 1983 (28. 04. 83) (Family : none)	1 - 16, 19 - 20
Y	JP, A, 55-139448 (Toray Industries, Inc.), 31 October 1980 (31. 10. 80), Page 4, upper right column, line 17 to page 4, lower left column, line 7 & EP, A, 17,942 & US, A, 4,284,540	1 - 20
Y	JP, A, 56-99248 (Toray Industries, Inc.), 10 August 1981 (10. 08. 81), Page 4, upper right column, line 13 to page 4, lower left column, line 3 (Family : none)	1 - 4, 6 - 20

V. ☐ OBSERVATIONS WHERE CERTAIN CLAIMS WERE FOUND UNSEARCHABLE[10]

This international search report has not been established in respect of certain claims under Article 17(2) (a) for the following reasons:

1. ☐ Claim numbers_____ because they relate to subject matter[12] not required to be searched by this Authority, namely:

2. ☐ Claim numbers_____ because they relate to parts of the international application that do not comply with the prescribed requirements to such an extent that no meaningful international search can be carried out[13], specifically:

VI. ☐ OBSERVATIONS WHERE UNITY OF INVENTION IS LACKING[11]

This International Searching Authority found multiple inventions in this international application as follows:

1. ☐ As all required additional search fees were timely paid by the applicant, this international search report covers all searchable claims of the international application.

2. ☐ As only some of the required additional search fees were timely paid by the applicant, this international search report covers only those claims of the international application for which fees were paid, specifically claims:

3. ☐ No required additional search fees were timely paid by the applicant. Consequently, this international search report is restricted to the invention first mentioned in the claims; it is covered by claim numbers:

4. ☐ As all searchable claims could be searched without effort justifying an additional fee, the International Searching Authority did not invite payment of any additional fee.

Remark on Protest

☐ The additional search fees were accompanied by applicant's protest.

☐ No protest accompanied the payment of additional search fees.

FURTHER INFORMATION CONTINUED FROM THE SEC ND SHEET

Y	JP, A, 57-34152 (Toray Industries, Inc.) 24 Feburary 1982 (24. 02. 82), Page 4, lower left column, line 11 to page 4, lower right column, line 2 (Family : none)	1 - 20
Y	JP, A, 57-192454 (Toray Industries, Inc.), 26 November 1982 (26. 11. 82), Page 3, lower left column, lines 1 to 11 (Family : none)	1 - 20
Y	JP, A, 56-30460 (Toray Industries, Inc.) 27 March 1981 (27. 03. 81), Page 4, upper right column, lines 8 to 20 (Family : none)	1 - 20

V. ☐ OBSERVATIONS WHERE CERTAIN CLAIMS WERE FOUND UNSEARCHABLE[10]

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Remark on Protest

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☐ No protest accompanied the payment of additional search fees.

FURTHER INFORMATION CONTINUED FROM THE SECOND SHEET

Y	JP, A, 56-159247 (Toray Industries, Inc.), 8 December 1981 (08. 12. 81), Page 3, lower right column, lines 5 to 16 (Family : none)	1 - 20
Y	JP, B, 58-47419 (Toray Industries, Inc.) 22 October 1983 (22. 10. 83) (Family : none)	1 - 20
Y	JP, A, 58-171438 (Toray Industries, Inc.), 8 October 1983 (08. 10. 83) (Family : none)	1 - 20
Y	JP, A, 55-86835 (Toyobo Co., Ltd.), 1 July 1980 (01. 07. 80), Page 3, lower left column, line 17 to page 3, lower light column, line 3	1 - 4, 6 - 20

V. ☐ OBSERVATIONS WHERE CERTAIN CLAIMS WERE FOUND UNSEARCHABLE [10]

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Remark on Protest

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International Application No. PCT/JP85/00242

FURTHER INFORMATION CONTINUED FROM THE SECOND SHEET

	(Family : none)	
Y	JP, A, 58-45253 (Teijin Ltd.), 16 March 1983 (16. 03. 83) (Family : none)	13 - 16
Y	JP, A, 57-125253 (Sumitomo Chemical Co., Ltd.), 4 August 1982 (04. 08. 82) (Family : none)	1 - 20
Y	JP, A, 58-201842 (Toray Industries, Inc.), 24 November 1983 (24. 11. 83) (Family : none)	1 - 20
Y	JP, B, 57-26303 (Teijin Ltd.), 3 June 1982 (03. 06. 82) (Family : none)	1 - 20

V. ☐ OBSERVATIONS WHERE CERTAIN CLAIMS WERE FOUND UNSEARCHABLE[10]

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Remark on Protest

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☐ No protest accompanied the payment of additional search fees.

FURTHER INFORMATION CONTINUED FR M THE SECOND SHEET

Y	JP, A, 51-89558 (Teijin Ltd.), 5 August 1976 (05. 08. 76) (Family : none)	1 - 20
Y	JP, A, 51-39749 (Mitsubishi Gas Chemical Co., Inc.), 2 April 1976 (02. 04. 76) (Family : none)	1 - 20
Y	JP, A, 54-91557 (General Electric Company), 20 July 1979 (20. 07. 79) & DE, A, 2,757,557 & NL, A, 7,714,377 & AU, A, 31,196,777 & FR, B, 2,375,312	1 - 20
Y	JP, A, 58-17150 (Toray Industries, Inc.), 1 February 1983 (01. 02. 83) (Family : none)	1 - 20

V. ☐ OBSERVATIONS WHERE CERTAIN CLAIMS WERE FOUND UNSEARCHABLE[10]

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Remark on Protest

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☐ No protest accompanied the payment of additional search fees.

FURTHER INF RMATION C NTINUED FROM THE SECOND SHEET		
Y	JP, A, 54-83053 (Teijin Ltd.), 2 July 1979 (02. 07. 79) (Family : none)	1 - 20

V. ☐ BSERVATIONS WHERE CERTAIN CLAIMS WERE FOUND UNSEARCHABLE [10]

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